FINAL REPORT

Title: Residual Stress Strengthened Functionally Gradient Materials
Grant Number: DMR-9400261
Investigators: Karl Jakus and John E. Ritter
Amount: $585,000
Period: 8/1/94 - 7/31/99

Summary of Results

The principal objective of this research was to develop a fundamental understanding of how residual stresses can be used to strengthen ceramic materials. An additional objective was to develop novel processing techniques to impart beneficial residual stresses to ceramic component. To achieve these objectives research was conducted in six thrust areas. Four of these areas relate to residual stress generation. The fifth area is related to an experimental technique to measure surface residual stresses, whereas the sixth area is focused on the effect of elastic modulus gradients on the contact-damage resistance of ceramic components. The six thrust areas were:

1. Contact Creep
2. Localized Creep
3. Non-uniform Pressure Assisted Densification
4. Creep Load Transfer in Fiber Reinforced Composites
5. Nano-indentation to Measure Residual Surface Stresses
6. Effect of Elastic Modulus Gradient on Contact-damage Resistance

Research in all six areas produced interesting and valuable results that are documented in publications acknowledging the support of this grant. A brief description of the accomplishments in each of the thrust areas is given in the following.

<u>Contact Creep</u>

When a ceramic material creeps under localized contact pressure and is allowed to cool under load a portion of the creep flow stress is retained as a residual stress due to constraints in elastic rebound. Generating such localized compressive residual stress at critical areas of ceramic components could result in an overall improvement of strength and reliability. To evaluate the concept of contact creep for residual stress generation a 2mm x 2.6mm rectangular flat indenter was pressed into the surface of three different materials at different pressures and temperatures, namely soda-lime glass, a 95% purity vitreous bonded alumina, and 99.5% high purity alumina. The resulting residual stress at the impression site was measured by Vickers indentation. Finite element calculations were also made for the expected contact residual stress. Residual compressive stresses were found within the contact area and corresponding tensile stresses outside the contact area. The results of this research have been published in the Journal of Materials Research.

<u>Localized Creep</u>

When a ceramic component is put under service load at high temperature certain regions of the component will experience creep deformation. Then, if the component is cooled under load, the tensile creep regions will develop compressive residual stresses due to constraints in elastic rebound. It is expected that such residual compression will have a beneficial effect on the overall reliability of the component.

To evaluate the concept of strengthening by localized creep single edge notched flexure specimens of soda-lime glass, alumina, and hot-pressed silicon nitride were crept at high temperature and unloaded at room temperature. Comparing the before-creep and after-creep strength distribution of the specimens assessed residual stress generation. Although, finite element calculations did not predict residual stress generation for soda-lime glass, experimental results showed statistically significant strengthening that depended on both the creep flow stress and the duration of creep. It is argued that the strengthening in this case was the result of creep induced crack blunting at the notch tip rather than residual compressive stress.

Finite element calculations indicated that the amount of residual stress depends on the creep stress exponent. Larger stress exponent results in larger retained stress. Therefore, it was expected that such materials as alumina and silicon nitride with tensile creep stress exponents in the range of 2 to 4 would exhibit significant strengthening by localized creep. Unfortunately, results to date did not bear out this prediction. For alumina any residual stress strengthening was obscured by creep damage. Strengthening was observed for silicon nitride; however, it was primarily due to previously reported oxidation effects.

The results of this research were presented at the Cocoa Beach meeting of the Engineering Ceramics Division of ACS in January 1996 and a corresponding paper appeared in the Ceramics Engineering and Science Proceedings.

<u>Non-uniform Pressure Assisted Densification</u>

A novel aspect of the originally proposed research was the generation of residual stress by hot pressing or HIP-ing ceramic compacts with spatial gradients in their properties such as green density or composition. Property gradients are expected to result in stress gradients that lead to residual stresses in the densified component. For example, higher density regions of a compact would require higher stress to densify at a given rate than lower density regions. Cooling the compact under load and unloading at room temperature can capture these stress differences. The stresses would be redistributed by elastic rebound such that residual compression would be balanced by residual tension. Thus, it is believed that judiciously designed property gradients could lead to beneficial residual stresses. For example a narrow compression layer on the surface of a component would be balanced by a lower magnitude tension in the interior. This strengthening by surface compression is similar to that based on differential thermal expansion except in this case the residual stresses would not diminish with increasing temperature. Prior to this research no one has proposed strengthening by non-uniform densification nor has anyone studied stress evolution during pressure assisted sintering from the point of view of controlling residual stresses.

In order to predict densification and stress evolution during hot pressing or HIP-ing compacts with property gradients, a densification model is needed that is applicable for the full range of densities. Existing models were derived for either low or high densities. The low-density models assume that the porosity is open and interconnected; whereas the high-density models assume that the pores are all closed. It was found that the low and high-density models are not compatible with each other in the presence of deviatoric (shear) stresses. Namely, discontinuity results in the predicted densification rate as well as stress evolution at the point of switching from one model to the other. This is not surprising since the implication that during densification all pores would remain open up to a certain density and then, suddenly, they would all become closed for the rest of the densification process. Such behavior is unlikely in nature. Because of the incompatibility, the existing models cannot be used separately to predict densification of compacts with property gradients since these compacts can develop significant deviatoric stresses. It is important to be able to predict densification of heterogeneous bodies not only for the present case of controlled residual stress generation but also for the case of hot pressing and HIP-ing functionally gradient ceramic materials that received considerable attention during the recent past.

To eliminate discontinuities in the prediction of densification rate and stress, a unified model has been developed that blends the low and high-density models from the onset of densification. The physical argument for this is that the average porosity in the compact is likely to shifts gradually from open to closed character with increasing density. As a result of the gradual translation from one model to the other, predictions are free of discontinuities and the model can be used to analyze compacts with spatial variation of properties. In addition to eliminating discontinuities, the effects of pressureless sintering have also been incorporated into the unified model. It was felt that densification with property gradients would cause some regions of the compact to be under relatively low stress where densification would be driven by internal rather than external forces. The resulting unified model was implemented as a subroutine into a commercial finite element code (Abacus) in order to analyze the densification of compacts with complex geometry.

The material constants that appear in the model were evaluated by fitting the predictions to experimental data. Homogeneous alumina compacts were hot pressed with different pressures at different temperatures. The resulting data enabled the evaluation of creep stress and activation energy constants that play a role in the model. Subsequent predictions of densification of alumina compacts with green density gradients were made using these constants.

Predictions for cylindrical compacts with a variety of radial green density gradients revealed the subtle evolution of density and stress during hot pressing. For example, pressing with a constant force tends to diminish the initial gradients in density and stress as densification proceeds. However, pressing with a constant displacement rate amplifies the initial gradients. Such processing path sensitivity is the result of the non-linear dependence of the densification rate on pressure and the synergistic interaction between creep deformation and densification. These results are significant not only for the residual stress-strengthening problem but also for predicting undesirable residual stress evolution in conventional hot pressing and HIP-ing.

The densification model was also extended to the mixtures of alumina and zirconia powders. A small amount of zirconia addition to alumina powder has been shown to drastically reduce the densification rate. This behavior can be used to design alumina components with a thin layer zirconia doped alumina such that upon hot-pressing a surface compressive layer would be produced. Experimental results clearly showed the effects of zirconia addition. Hot-pressed cylindrical composites developed excessive amount of tensile stresses at the ends and, as a consequence, they suffered cracking. Further work remains to optimize the zirconia doping level and the relative thickness of the layers of different compositions.

The results of the non-uniform densification thrust have been published in Ceramics Engineering and Science Proceedings, Journal of Material Science, and the Journal of the American Ceramic Society.

Creep Load Transfer in Fiber Reinforced Composites

Another thrust area that represents an innovative approach to strengthening ceramics involves high temperature load transfer between the matrix and the reinforcing fibers of ceramic matrix composites. It was postulated that if the applied load is allowed to transfer by creep from the matrix to the fibers, and if this composite is cooled under load, the fibers would pull the matrix into compression upon unloading. This compressive residual stress would effectively increase the first-matrix-cracking stress that is often the design stress for ceramic composites. This method of strengthening is analogous to the strengthening of pre-stressed concrete. A demonstration of this concept was undertaken using Nicalon fiber reinforced BMAS (boron doped magnesium aluminum silicate) glass-ceramic matrix composite. The material was obtained from Dr. John Brennan of United Technologies who was the developer of this material. The practical objective of this study was to demonstrate that high temperature creep could increase the first matrix cracking stress. The fundamental objective was to gain an understanding of the creep load transfer mechanism in ceramic composites. This is an important issue not only for the practical goal of strengthening but also for predicting mechanical property shifts in high temperature applications. As the relative load carried by the fibers and the matrix shifts due to creep, the strength and reliability of the composite might become compromised.

Load transfer experiments with the Nicalon/BMAS composite were carried out at the High Temperature Materials Laboratory (HTML) of Oak Ridge National Laboratory (ORNL). One of our graduate students did the work under the supervision of Dr. Edgar Lara-Curzio. Specimens were loaded in uni-axial tension at 1100^{o}C under different loads for different lengths of time. Following the load transfer treatment, the specimens were tensile tested at room temperature. During testing an acoustic emission sensor monitored cracking activity. For a few specimens the residual matrix stress was also measured by x-ray diffraction.

The results of this research are very encouraging. Increases in first-matrix-cracking stress to more than two times the as-received value has been achieved without affecting either the un-cracked elastic modulus, or the ultimate strength, or the strain to failure of the composite. The onset of matrix cracking detected by acoustic emission correlated with the proportional limit in the room temperature mechanical tests. It is believed that the predominant cause for the increased first-matrix-cracking stress (or proportional limit) is residual compression in the matrix due to creep load transfer. The x-ray diffraction results confirm the existence of residual compression in the matrix.

This thrust area provided an excellent opportunity for the UMass researchers to collaborate with scientists in other institutions. Namely, Dr. Brennan (composite developer) of United Technologies, Drs. Lara-Curzio (mechanical behavior expert), Sun (Nicalon/BMAS characterization expert), Watkins (x-ray diffraction expert) of ORNL was all intimately involved in the project. The graduate student participant gained valuable experience by having an opportunity to work with these scientists and by learning to use state of the art instrumentation.

One of the most intriguing aspects of the load transfer results is the increase of first-matrix-cracking stress to a level above the applied creep stress. This is counter-intuitive if all the increase is due to load transfer. Something, in addition to load transfer, is going on but several different experiments designed to resolve this question did not provide a satisfactory answer. Perhaps the cause is related to the complex geometry of the fiber layout (eight plies in $0/90^o$ orientation). The answer will have to be obtained from a systematic study of the high temperature load transfer mechanism.

The results of the creep load transfer thrust have been published in Ceramics Engineering and Science Proceedings, Journal of the American Ceramic Society, ASME conference proceedings.

Nano-indentation to Measure Residual Surface Stresses

Nano-indentation has emerged in recent years as a powerful tool to study the mechanical properties of metals and ceramics on a microscopic scale. The cracks that form in glasses and ceramics as a result of nano-indentation are a function of the material toughness and the local residual stress. The feasibility of using nano-indentation induced crack lengths to measure residual stresses in silicon nitride was evaluated in this research. Hot-pressed silicon nitride bend bars were nano-indented in-situ a four-point bend fixture at the nano-indentation facility of Oak Ridge National Laboratory. The resulting crack lengths were correlated with calculated local elastic stresses in the beam. The results showed excellent statistical correlation indicating that nano-indentation can be a viable tool to measure surface residual stresses in those cases where multiple indentations can be made under the same local conditions.

The results of the nano-indentation thrust were published in the proceedings of the CIMTEC98 conference held in Florence, Italy.

Effect of Elastic Modulus Gradient on Contact-damage Resistance

Previous research has shown that Hertzian cone cracking is suppressed in ceramic composites whose elastic modulus increases with depth below the surface. The objective of this research thrust was to determine if these modulus-graded composites would also exhibit superior resistance to particle impact damage. Therefore, impact damage with sharp and blunt particles was studied in monolithic and modulus-graded glassy alumina. The composite was fabricated by impregnating a dense, fine-grained alumina with an aluminosilicate glass having a lower elastic modulus than the alumina. This produced a functionally gradient composite with decreasing glass content below the surface, thus causing the elastic modulus to monotonically increase with depth. The aluminosilicate glass had a coefficient of thermal expansion and Poisson ratio the same as the alumina. Therefore, the composite had no macroscopic, long-range residual stresses.

The sharp multi-particle impact (erosion) experiments were conducted with a slinger type apparatus. The blunt single impact experiments were conducted with small stainless steel and tungsten carbide balls using a particle accelerator gas-gun. In the case of the sharp particle impact experiments, erosive wear and strength degradation was measured as a function of depth below the surface. For the single impact experiments, the onset of ring cracking was determined and compared to that for monolithic alumina. It was found that the modulus-graded alumina exhibited the same erosive wear by sharp particles and post erosion strength as monolithic alumina. For the spherical impact part of the research it was found that modulus gradation improved the impact damage resistance when high elastic modulus tungsten carbide projectiles were used. No improvement was observed using the lower elastic modulus chrome steel projectiles. Monolithic alumina impacted with tungsten carbide projectiles followed the predictions of impact theory and agreed reasonably well with the previously published quasi-static data. However, modulus-graded alumina impacted with tungsten carbide projectiles and both monolithic and modulus-graded alumina impacted with steel projectiles did not follow either the theory or the previous data. It is believed that the low modulus steel deforms more upon impact than the alumina specimen, thus overshadows the effects of the modulus gradient. On the other hand, the high modulus tungsten carbide does not deform appreciably during impact, allowing the modulus gradient to play a role in reducing the impact stresses and increasing the cracking threshold.

The results of this research thrust were published in Ceramics Engineering and Science Proceedings and the proceedings of a functionally graded materials symposium.

Students supported by the Grant

The grant supported both graduate and undergraduate students. Six of the eight graduate students who have earned their advanced degrees under this NSF grant found employment in industry. One has an academic position at the University of Singapore. The undergraduate students who were involved in the research were working side by side with the graduate students on projects for which they had individual responsibility.

Graduate:		Undergraduate:
James Webb	PhD, 1995	Robert Jensen
Revti Atri	MS, 1996	Robert Skolnik
Matt Westort	MS, 1996	Ileana Miranda
Sujanto Widjaja	PhD, 1997	Todd Emmonds
Sundeepan Batthacharya	PhD, 1997	Marc Leverdier
Rahul Panat	MS, 1999	Carlos Elguero
Kabir Bhatia	MS, 1999	Fitzroy Ward-Cawdette

Premal Shah	MS, 2000	Juan-Pablo Mas

Publications during the Grant period

S. Widjaja, K. Jakus, and J. E. Ritter, "Hertzian Indentation Damage in Alumina with R-Curve Behavior," *Ceram. Engr. Sci. Proc.* **16** [4] 579-86 (1995).

J. E. Webb, K. Jakus, and J. E. Ritter, "Slow Crack Growth in Alumina with R-Curve at Elevated Temperatures," *Ceram. Engr. Sci. Proc.*, **16** [4] 571-8 (1995).

S. Widjaja, J. E. Ritter, & K. Jakus, "Influence of R-Curve Behavior on Strength Degradation Due to Hertzian Indentation," *J. Mat. Sci.*, **31**, 2379-84 (1996).

J. E. Webb, K. Jakus, and J. E. Ritter, "R-Curve and Subcritical Crack Growth Behavior at Elevated Temperatures in Coarse Grained Alumina," *Acta Metallurgica et Materialia*, **44**, 2259-66 (1996).

K. Jakus, J. E. Ritter, R. Atri, and S. Widjaja, "Creep-Induced Strengthening of Soda-Lime Glass and Silicon Nitride," *Ceram. Engr. Sci. Proc.*, **17** [4] 421-9 (1996).

J. E. Webb, K. Jakus, and J. E. Ritter, "High Temperature Bridging in Vitreous Alumina," *J. Am. Ceram. Soc.*, **80** [2] 509-12 (1997).

S. Widjaja, K. Jakus, R. Atri, J. E. Ritter, and S. Bhattacharya, "Residual Surface Stress by Localized Contact Creep," *J. Mat. Res.*, **12** [1] 210-17 (1997).

S. Bhattacharya, K. Jakus, and I. Grosse, "Hot Pressing Analysis of Ceramic Powders with Density Gradients," *Ceram. Engr. Sci. Proc.*, **18** [3] Part B, 3-10 (1997).

S. Widjaja, K. Jakus, J. E. Ritter, E. Lara-Curzio, E. Y. Sun, T. R. Watkins, and J. J. Brennan, "High-Temperature Load Transfer in Nicalon/BMAS Glass-Ceramic Matrix Composites," *Ceram. Engr. Sci. Proc.*, **18** [3] Part A, 681-88 (1997).

S. Bhattacharya, K. Jakus, and I. Grosse, "Modeling Pressure Assisted Densification by Power Law Creep," *J. Mat. Sci*., **32**, 6183-89 (1997).

S. Bhattacharya and K. Jakus, "Hot Pressing of Annular Alumina/Zirconia Composites," *J. Am. Ceram. Soc*., **81** [3] (1998).

K. Jakus, N. Evans, and J. Hay, "Stress Measurement with Nano-Indentation," pp 691-96, Proc. of 9th Int. Conf. On Modern Materials and Technologies (CIMTEC'98), Florence, Italy, June 14-19, 1998.

K. Jakus, S. Widjaja, and J. E. Ritter, "Correlation of Acoustic Emission with Stress-Strain Behavior in Fiber Reinforced Ceramic Matrix Composites," pp 35-42, Proc. 1997 ASME Int. Mechanical Engr. Congress and Exposition, "Recent Developments in Fracture Sensing and Analysis," AMD 971 Dallas, TX, November 16-21, 1997.

S. Widjaja, K. Jakus, J. E. Ritter, E. Lara-Curzio, E. Y. Sun, T. R. Watkins, and J. J. Brennan, "Creep-Induced Residual Stress Strengthening in a Nicalon Fiber-Reinforced BMAS Glass Ceramic Matrix Composite," *J. Am. Ceram. Soc*., **82** [3] pp 657-64 (1999).

K. Bhatia, K. Jakus, and J. E. Ritter, "Pressure Assisted Sintering of Mixtures of Alumina/Zirconia Powders," *Ceram. Engr. Sci. Proc.*, **20**, (1999).

R. P. Panat, K. Jakus, J. E. Ritter, and P. Shah, "Erosion and Strength Degradation of an Elastic Modulus Graded Alumina-Glass Composite," *Ceram. Engr. Sci. Proc.*, **21** [3] pp 635-42 (2000).

P. Shah, K. Jakus, and J. E. Ritter, "Impact Damage in Monolithic and Functionally Graded Alumina," to appear in Proc. FGM2000, 6th Int. Symp. on Functionally Graded Materials, Estes Park, CO., Sept. 10-13, 2000.